Spontaneous emission of non-dispersive Rydberg wave packets

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Non dispersive electronic Rydberg wave packets may be created in atoms illuminated by a microwave field of circular polarization. We discuss the spontaneous emission from such states and show that the elastic incoherent component (occuring at the frequency of the driving field) dominates the spectrum in the semiclassical limit, contrary to earlier predictions. We calculate the frequencies of single photon emissions and the associated rates in the "harmonic approximation", i.e. when the wave packet has approximately a Gaussian shape. The results agree well with exact quantum mechanical calculations, which validates the analytical approach.

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I. INTRODUCTION

Recent theoretical studies predicted the existence of non-dispersive wave packets in atoms driven by a microwave radiation of either circular [1] or linear polarization [2]. Contrary to standard electronic wave packets which disperse in time when the binding force is nonlinear, these novel objects do not spread due to a nonlinear coupling between the atom and the driving field which locks the electronic motion onto the driving frequency. The phenomenon responsible for this locking is a 1:1 resonance between the internal (Kepler) and external (microwave) frequencies, creating a resonance island at the center of which lies a stable periodic orbit. In the dressed atom picture [3], a single eigenstate of the full "dressed" hamiltonian represents the wave packet [2,4], explaining immediately the absence of spreading.

The very same microwave field is also responsible for the decay of the wave packet via ionization. As shown elsewhere [5], the typical ionization time of wave packets built from circular Rydberg states around principal quantum number $n_0 = 60$ exceeds 10^5 Kepler periods being often longer than 10^6 periods. The ionization occurs via a "chaos assisted tunnelling" mechanism [6] (the tunneling occurring from the stable island into the chaotic sea surrounding it). In effect the lifetime against ionization is very sensitive to small changes of the parameters (microwave frequency, Ω or its amplitude F) exhibiting large scale fluctuations [6].

Another possible decay mechanism of the wave packet is of course the spontaneous emission of photons. For circularly polarized microwave, the situation considered in this paper, it has been discussed in [7]. Here we readdress this issue since, in our opinion, the results reported in [7] are incomplete. To this end, we use a different approach, borrowing heavily from the well known treatment of resonance fluorescence of two-level atoms driven by a laser field [3] using the dressed atom approach. The main advantage of the present approach is its generality: it can be used for any polarization of the microwave field, for any superposition of driving fields with various frequencies and for any structure of electromagnetic modes surrounding the atom (as shown e.g. in [8], the presence of a cavity can strongly affect the spontaneous emission of an atom). Also, it uses the common knowledge widely developed in the quantum optics community and is thus accessible to a large audience. Let us mention also that spontaneous emission of non-dispersive wave packets in linearly polarized microwave is discussed in [9].

The paper is organized as follows. In section II, we discuss the basic properties of the non-dispersive wave packets (not considering spontaneous emission) from different points of view: laboratory frame or rotating frame, time-dependent or Floquet pictures, dressed atom picture. The discussion is necessary for a proper understanding of spontaneous emission, which is done in section III. Our results are compared to the earlier published results in section IV. Section V is devoted to the validity of the harmonic approximation, by comparing our analytical results to "exact" numerical calculations. We conclude in section VI.

II. THE NON-DISPERSIVE WAVE PACKETS FROM DIFFERENT POINTS OF VIEW

We consider a hydrogen atom in the presence of a monochromatic driving microwave field of circular polarization, that is an electromagnetic field which can be described by a single mode. In this section, we will *not* consider any coupling to other (initially empty) modes of the electromagnetic field. This will be discussed in section III. We first treat the microwave field classically. A full quantum treatment of the microwave field is possible (see section II D), but not really needed as, for usual microwave fields, the number of photons in the mode is enormous and a classical treatment of the microwave field is possible.

A. Hamiltonian in the laboratory frame

With the previous assumption, the quantum Hamiltonian of the system is (in atomic units, neglecting relativistic and QED effects, assuming an infinite mass of the nucleus and in the dipole approximation):

$$H_{\text{cl}} = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + F(x \cos \Omega t + y \sin \Omega t). \quad (1)$$

where Ω is the frequency of the microwave field circularly polarized in the $x - y$ plane and F its amplitude. The x-axis is chosen to coincide with the microwave electric field at $t = 0$.

B. Floquet approach

The Hamiltonian (1) is periodic in time with period $2\pi/\Omega$. Hence, a Floquet approach [10] may be used: any solution of the time-dependent Schrödinger equation can be expanded as a linear combination of *time-periodic* states - called Floquet states of the system - with coefficients oscillating in time as $\exp(-iE_i t)$ where E_i is called the quasi-energy of the Floquet state (there is no explicit $\hbar$ in the equations as we are using atomic units where it is one). Obviously, the quasi-energy is defined only modulo Ω making the Floquet spectrum periodic in energy with period precisely Ω. The Floquet spectrum as well as the Floquet states can be obtained from the diagonalization of a suitable operator, namely the Floquet Hamiltonian:

$$\mathcal{H} = H_{\text{cl}} - i \frac{\partial}{\partial t} \quad (2)$$

with time-periodic boundary conditions. A suitable basis for the Floquet Hamiltonian is the product of any basis in usual space - for example, the usual hydrogenic eigenbasis with bound states $|nlm\rangle$ - by the basis of time-periodic oscillating exponentials $\exp(iK\Omega t)$ with K an integer. We will denote $|nlm; K\rangle$ such a basis. The next step is to expand the time-periodic Hamiltonian in a Fourier series and calculate the non-zero matrix elements of the Floquet Hamiltonian. Because no higher harmonics of the microwave frequency exist in the Hamiltonian, only matrix elements changing K by zero or one unit do not vanish. Moreover, in the specific case of circular polarization, the part in the Hamiltonian which increases (resp. decreases) K by one is proportional to $x - iy$ (resp. $x + iy$) hence decreases (resp. increases) the angular momentum of the electron along the z axis by one unit. In other words, the Floquet Hamiltonian obeys the selection rule $\Delta(K + m) = 0$. This signifies merely that absorption of a σ^+ polarized microwave photon must increase the atomic angular momentum $L_z = xp_y - yp_x$ by one unit. The Floquet Hamiltonian thus has a block diagonal structure and the Hilbert space can be split into manifolds characterized by constant value of $\mathcal{K} = m + K$. In each block, the

Floquet Hamiltonian is no longer periodic in energy. On the other hand, the Floquet Hamiltonian in blocks $\mathcal{K}$ and $\mathcal{K}'$ only differ by a constant energy term $(\mathcal{K}' - \mathcal{K})\Omega$ which restores the total periodicity of the Floquet spectrum. However, diagonalization in a given block is sufficient to obtain all the information on the system.

C. Hamiltonian in the rotating frame

For a circularly polarized radiation, another possibility exists which has been frequently used in the past [1,4–7,11–21]. By passing to the non inertial frame rotating with frequency Ω using the unitary transformation $U = \exp(i\Omega L_z t)$, one may remove the oscillatory time dependence from Eq. (1) obtaining in the rotating frame the Hamiltonian

$$H_{\text{rot}} = U \, H_{\text{cl}} \, U^\dagger + iU \frac{\partial U^\dagger}{\partial t} = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + Fx - \Omega L_z. \quad (3)$$

This time-independent Hamiltonian has some energy levels and corresponding eigenstates. Obviously, its spectrum is not Ω-periodic. However, the link with the Floquet approach in the laboratory frame is easily established: there is a one-to-one correspondence between the eigenstates in the rotating frame and the Floquet states in the laboratory frame from the block $\mathcal{K} = 0$. Indeed, consider an eigenstate (i.e. time-independent state) $|\phi_i\rangle$ of H_{rot} with energy E_i. From Eq. (3), we deduce straightforwardly:

$$\left(H_{\text{cl}} - i \frac{\partial}{\partial t}\right) U^\dagger |\phi_i\rangle = E_i U^\dagger |\phi_i\rangle \quad (4)$$

which implies (see Eq. (2)) that $U^\dagger |\phi_i\rangle$ is a Floquet eigenstate with quasi-energy E_i. From the definition $U^\dagger = \exp(-i\Omega L_z t)$, it is clear that the K-th Fourier component of $U^\dagger |\phi_i\rangle$ is nothing but the component of $|\phi_i\rangle$ on the subspace with $L_z = m = -K$. In other words, $U^\dagger |\phi_i\rangle$ is a Floquet eigenstate in the laboratory frame lying in the block $\mathcal{K} = m + K = 0$.

Of course, Floquet states belonging to the other blocks $\mathcal{K} \neq 0$ can also be found from the time-independent Hamiltonian in rotating frame. A first method is to use, instead of the operator U, a slightly different unitary equivalent operator $\exp(-ik\Omega t)U$ (with k an integer, it has the same time-periodicity as U itself) which leads to the following Hamiltonian in the rotating frame:

$$H_{\text{rot}} = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + Fx - \Omega(L_z - k). \quad (5)$$

Taking all possible positive and negative integer k values one recovers the full Floquet spectrum.

A second equivalent method is to keep the standard Hamiltonian in rotating frame, Eq. (3), and to consider states of the form $\exp(ik\Omega t)U^\dagger |\phi_i\rangle$. These are time-periodic Floquet (if k is an integer), eigenstates of the

Floquet Hamiltonian $\mathcal{H}$, Eq. (2) with energy $E_i + k\Omega$, belonging to the block $\mathcal{K} = m + K = k$.

At a deeper level, this can be seen as a consequence of the unitary equivalence (which implies the same spectrum) for the Floquet Hamiltonians in the laboratory frame and in the rotating frame. The spectrum of the latter is here very simple: because H_{rot} is time-independent, it is simply the energy spectrum of H_{rot} shifted by an arbitrary integer multiple of Ω. For a more complicated case like – for example – a microwave with polarization close to circular, the Hamiltonian in the rotating frame is weakly time-dependent and its corresponding Floquet Hamiltonian (with the time dependent term treated perturbatively) may be the simplest approach.

D. Dressed atom approach

An alternative view of this problem is to treat the microwave field quantum-mechanically as being in either a coherent state or a pure Fock state with a large number of photons in the circularly polarized mode [22]. This approach is quite common in quantum optics and is known as the dressed atom picture. Then the atom is not considered as an isolated quantum system, but it is rather the strongly coupled atom+field system which is treated using quantum mechanics. The Hamiltonian of the atom+field system is:

$$H_{\mathrm{q}} = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + V(d_- a^\dagger + d_+ a) + \Omega a^\dagger a, \qquad (6)$$

where $a^\dagger/a$ are the creation/annihilation operators in the occupied, σ^+ circularly polarized mode, $d_\pm = (x \pm iy)/\sqrt{2}$ the corresponding components of the dipole moment operator and V is the coupling strength, whose exact expression is given in section III A but not important at the present level.

The Hamiltonian H_{q} is time independent and may be expanded in the basis: $\{|nlm\rangle \otimes |N\rangle = |nlm; N\rangle\}$ where $|nlm\rangle$ are hydrogenic states and N denotes the number of microwave photons in the occupied mode. The diagonalization of the Hamiltonian yields then the dressed states; as its stands, the energy spectrum is not Ω-periodic.

The absorption of a circularly polarized microwave photon increases by one the projection L_z of the electron angular momentum on the z axis. In the quantum field basis this manifests itself in the fact that the Hamiltonian matrix is block diagonal. The Hilbert space splits into manifolds characterized by the total atom+field projection of the angular momentum $\mathcal{N}$, i.e., basis states coupled within a given block have fixed $\mathcal{N} = m + N$. $\mathcal{N}$ is simply the total angular momentum projection on the z axis carried by both the electron and the photons.

When the number of photons is very large, $N \gg 1$, we can neglect the variations in the non-diagonal matrix elements – proportional to either $\sqrt{N}$ or $\sqrt{N+1}$ – across neighboring N blocks. Then, $\sqrt{2}V\sqrt{N}$ can be identified

as the amplitude of the microwave field F. With this approximation, the energy spectrum is (locally) Ω-periodic. As a matter of fact, the matrix elements of H_{q} are obviously equal to the ones of the classical Floquet Hamiltonian, Eq. (2), already discussed in section II B, provided that the quantum number K labelling the Fourier components in the Floquet picture is identified with the number of photons N. This establishes – in the limit of a large number of photons – the one-to-one correspondence between the quasi-energy spectrum of the Floquet Hamiltonian and the energy spectrum of the dressed atom, and also the correspondence between the Floquet eigenstates and the dressed atom eigenstates. The two points of view are completely equivalent: in the following, we will use the dressed atom picture as it seems more convenient for incorporating the spontaneous emission of photons.

As a final remark, let us note that the previous discussion is already well known in a different context in quantum optics: for the simple case of a two level atom coupled resonantly by an oscillatory field, in the rotating wave approximation. There, a field-quantized or Floquet approach yields an energy spectrum periodic in Ω – a sequence of doublets separated by the Rabi frequency [3] – while semiclassically in the "rotating frame" [23] one gets a single doublet of states.

E. Numerical calculation of the energy spectrum

As discussed in the three preceding sections, there exists a one-to-one correspondence between the eigenstates in the rotating frame, the Floquet eigenstates and the dressed states in the quantum language. Therefore, we shall call in the following the eigenenergies of either of the Hamiltonians (2),(3) or (6) as quasienergies while referring to the eigenstates as Floquet eigenstates or dressed states.

As we are interested in the regime of highly excited Rydberg states (with typical principal quantum number 60) driven by a resonant microwave field, the number of atomic states significantly coupled by the microwave field is enormous. For computational efficiency, it is better to use a Sturmian basis [4–6] rather than the usual atomic basis. The advantage is that it provides a discretization of the atomic continuum. Combined with the complex rotation technique, it allows us to find not only the energies of the dressed states but also their lifetimes against ionization by the microwave field [5,6]. To numerically find several converged dressed states, diagonalizations of very large matrices (of the order of 10^5) are necessary. As discussed above, there exists a full equivalence between diagonalization of the Floquet Hamiltonian, i.e. clever use of the time-periodic structure of the problem and diagonalization of the Hamiltonian in the rotating frame. The computational efficiencies of the two points of view are strictly equal. Our computer code [4–6] diagonalizes in fact H_{rot}, contrary to the statement in [19].

F. Non-dispersive wave packets

Some time ago, Klar [24] noticed that the Hamiltonian (3) in the rotating frame allows for the existence of a stable fixed (equilibrium) point in a certain range of microwave amplitude F. The beautiful contribution of [1] was to notice that wave packets initially localized in the vicinity of this fixed point will not disperse (being bound by the fact that the fixed point is stable) for very long time. In the laboratory frame, these wave packets also called "Trojan states" in [1] appear as wave packets moving around the nucleus along the circular trajectory [the periodic orbit of (1)]. The existence of "stationary" wave packets – keeping exactly their shape during propagation along the circular orbit and hence not dispersing at arbitrarily long times (neglecting the very slow ionization) – was later found in [4]. These are simply some among the eigenstates of the dressed atom: by construction, they are strictly periodic functions of time.

Semiclassically, the wave packets are manifestations of the motion inside the stable primary 1:1 resonance zone when the microwave frequency matches the electronic frequency. Similar states have been studied earlier for model one-dimensional driven systems [25]. While for other models, the wave packet resembles itself every period of the driving perturbation (while changing shape within one period [2,25]), the wave packets in circular polarization keep the same shape at all times. This is due to the conservation of $\mathcal{N}$.

Quantum mechanically, any dressed eigenstate may be viewed as a linear combination of atomic states, combined with appropriate photonic states (in such a way that in the combination $\mathcal{N}$ is conserved). The wave packet states are linear combinations of atomic Rydberg states coupled by resonant microwaves. Thus, the biggest contribution to this combination comes from circular states with principal quantum number n close to:

$$n_0 = \Omega^{-1/3}. \tag{7}$$

Note that n_0 is not necessarily an integer. For the particular case of the "ground state wave packet" (with maximum localization near the fixed point), it is a superposition of mainly circular atomic states $|n, n-1, n-1\rangle$ with a small contamination of other quasi-two-dimensional states of the form $|n, l, l\rangle$ and even smaller contributions from states $l \neq m$ extending significantly out of the xy plane.

The fixed point in the rotating frame is located at $y_e = z_e = 0$ and x_e satisfying

$$1/x_e^2 + F - x_e\Omega^2 = 0. \tag{8}$$

It is convenient to introduce [1] a dimensionless parameter:

$$q = \Omega^{-2}x_e^{-3}. \tag{9}$$

Then $q = 1$ for $F = 0$ and the fixed point is stable for $q \in [8/9, 1]$. It is also useful to define scaled variables which make the discussion of the results less dependent on the particular microwave frequency value [26]. We have already defined the effective principal quantum number n_0, Eq. (7). The scaled microwave amplitude is defined as

$$F_0 = Fn_0^4 = F\Omega^{-4/3}. \tag{10}$$

F_0 is a dimensionless parameter simply related to q, see Eq. (9), via

$$F_0 = (1 - q)q^{-1/3}. \tag{11}$$

A possible way for creation of a non-dispersive wave packet consists of the preparation of a circular state with a given principal quantum number n_0 followed by a smooth turn on of the microwave with frequency $\Omega = 1/n_0^3$ [4]. Such an approach has been tested numerically by integration of the time-dependent Schrödinger equation [27]. It has been shown that the wave packet may be obtained with about 90% efficiency.

During the switch-on of the microwave, two regimes may be identified. For q only slightly less than unity, (small microwave amplitude), the resonant coupling between circular states is important only. The effective approximate Hamiltonian in the vicinity of the fixed point corresponds then to the Hamiltonian of a pendulum [12]. This is the standard description of the resonance at first order [28]. For low F_0, the resonance island is small in action variable and, in effect, the pendulum states must be quite extended in the conjugate angle. In the rotating frame, this manifests itself as the fixed point being marginally stable along the y direction at vanishing F_0. For too small F_0, the binding force is too small and the wave packet eigenstate can be hardly considered to be localized in the very vicinity of the fixed point and, in turn, cannot be considered as a localized wave packet in the laboratory frame.

With increasing microwave amplitude, the resonance island grows and may support a few (depending on the size of the effective $\hbar = 1/n_0$) states. Then, for a sufficiently large F_0 (but not large enough to destabilize the resonance), the low excitations in the resonance island may be well described in the harmonic approximation. It is this regime of F_0 values which is of interest.

The harmonic expansion of the Hamiltonian $H_{\rm rot}$ in rotating frame around the equilibrium point allows one to find normal modes of the problem and obtain analytic predictions concerning the energies of wave packet eigenstates [1,4].

G. Harmonic approximation

The Hamiltonian $H_{\rm rot}$ can be expanded in powers of position and momentum operators in the vicinity of the

fixed point. The first order terms cancel because this is a fixed point. In the harmonic approximation, only second order terms are kept (including of course crossed position-momentum terms) and we obtain:

$$H_{\text{rot}} \approx H_{\text{h}} = E_e + \frac{\mathbf{p}^2}{2} + \Omega(\tilde{x}p_y - \tilde{y}p_x) + \frac{\Omega^2 q\tilde{y}^2}{2}$$
$$-\Omega^2 q\tilde{x}^2 + \frac{\Omega^2 q\tilde{z}^2}{2} \qquad (12)$$

where

$$E_e = \frac{1 - 4q}{2q^{2/3}}\Omega^{2/3} \qquad (13)$$

is the energy of the equilibrium point and $(\tilde{x}, \tilde{y}, \tilde{z}) = (x - x_e, y - y_e, z - z_e)$ denotes the displacement with respect to the fixed point.

The "diagonalization" of this Hamiltonian leads to the definition of the normal modes for the electronic motion. The calculation is tedious but straightforward: the frequencies of the 3 normal modes labelled by the $(+, -, z)$ indices are:

$$\omega_{\pm} = \Omega\sqrt{\frac{2 - q \pm Q}{2}}, \qquad (14)$$

$$\omega_z = \Omega\sqrt{q} \qquad (15)$$

with a shorthand notation

$$Q = \sqrt{9q^2 - 8q} \qquad (16)$$

The corresponding creation and annihilation operators in the 3 normal modes are linear combinations of position and momentum operators. In the harmonic approximation, the motion perpendicular to the plane of microwave polarization decouples from the motion in the plane; hence, the annihilation operator in the z-mode has the following simple expression:

$$b_z = \frac{q^{1/4}\sqrt{\Omega}\tilde{z} + iq^{1/4}p_z/\sqrt{\Omega}}{\sqrt{2}} \qquad (17)$$

The creation operator $b_z^{\dagger}$, being the adjoint of b_z, has a similar expression with imaginary part of opposite sign. The $\pm$ modes completely entangle the various coordinates and momenta $(\tilde{x}, \tilde{y}, p_x, p_y)$. They are neither the usual "cartesian" normal modes along $\tilde{x}$ and $\tilde{y}$, nor "circular" normal modes along $\tilde{x} \pm i\tilde{y}$, but intermediate combinations. The annihilation operators are:

$$b_{\pm} = N_{\pm}\left(\sqrt{\Omega}\tilde{x} - i\frac{(Q \pm 3q \mp 2)\Omega^{3/2}}{2\omega_{\pm}}\tilde{y}\right.$$
$$\left. -i\frac{(Q \pm q)\sqrt{\Omega}}{2q\omega_{\pm}}\tilde{p}_x + \frac{3q \pm Q}{2q\sqrt{\Omega}}\tilde{p}_y\right), \qquad (18)$$

with the normalization constants given by:

$$N_{\pm} = \left(\frac{q^2(2Q^2 - q + 2 \pm 3(1 - 2q)Q)}{8Q^2(1 - q)}\right)^{1/4}. \qquad (19)$$

The Hamiltonian in the harmonic approximation can be expressed in terms of creation and annihilation operators of the electronic normal modes [1,4,5]:

$$H_{\text{h}} = E_e + \omega_+(b_+^{\dagger}b_+ + 1/2)$$
$$-\omega_-(b_-^{\dagger}b_- + 1/2) + \omega_z(b_z^{\dagger}b_z + 1/2). \qquad (20)$$

Note the unusual minus sign in front of the ω_- term. This comes from the fact that the stable fixed point is not a minimum of the Hamiltonian. However, the presence of crossed momentum-position terms assures the complete stability of the equilibrium point versus any small perturbation, in complete analogy with a usual potential minimum.

Let us denote the atomic eigenstates in the harmonic approximation as $|n_+, n_-, n_z\rangle$ where n_i denotes the excitation in a given $\pm, z$ mode [4,5]. We will also – when needed – denote the Floquet or dressed atom eigenstates (thus including the microwave field degree of freedom) as $|n_+, n_-, n_z; \mathcal{N}\rangle$. The corresponding energies are:

$$E(n_+, n_-, n_z; \mathcal{N}) = E_e + \mathcal{N}\Omega + \left(n_+ + \frac{1}{2}\right)\omega_+$$
$$-\left(n_- + \frac{1}{2}\right)\omega_- + \left(n_z + \frac{1}{2}\right)\omega_z. \qquad (21)$$

It has been verified that exact Floquet quasienergies, corresponding to dressed states localized in the vicinity of the equilibrium point in the rotating frame agree with the harmonic prediction, Eq. (21), to within 10% of the mean level spacing [4,5] for wave packets corresponding to $n_0 \approx 60$ or higher. This indicates that the harmonic approximation is quite satisfactory for sufficiently large n_0 (sufficiently small Ω). In particular, the "ground state", $|0, 0, 0\rangle$ corresponds to the most tightly bound state, centered at the fixed point in the rotating frame. Its wavefunction has a Gaussian shape in the harmonic approximation. The corresponding exact Floquet (dressed) state [5] is a non-dispersive wave packet since in the laboratory frame it moves in time around the nucleus with frequency Ω following the stable circular periodic orbit. It is precisely the spontaneous emission from this state which will be discussed in the following sections.

The partial scheme of levels resulting from H_h is shown in Fig. 1 for a few values of the "total" angular momentum $\mathcal{N}$. The neighboring ladders of states are shifted with respect to each other by the microwave frequency Ω corresponding to different decoupled blocks of the Floquet Hamiltonian. Transitions between quasienergy levels (Floquet or dressed states) may be introduced by some additional perturbation. In particular one may consider the so-called Floquet spectroscopy [29], absorption or stimulated emission between dressed states introduced by an additional laser or microwave field.

It seems quite natural to study the possible effects of spontaneous emission of these states within the very same harmonic approximation. This allows to obtain close analytic expressions for the spontaneous decay rate. The

validity of the harmonic approximation has been challenged strongly by Farrelly and coworkers [16–20]. By comparison of spontaneous spectra obtained in the harmonic approximation with full quantum mechanical results, we show later that their claims are not justified.

III. SPONTANEOUS EMISSION OF THE WAVE PACKET IN THE DRESSED ATOM PICTURE

A. General properties

To consider the spontaneous emission one has to consider the atom interacting not only with a single mode [treated either classically (1) or quantum mechanically (6)] but a full electromagnetic field. The corresponding Hamiltonian reads, in atomic units [30]

$$H = H_i + \mathbf{r} \cdot \mathbf{E(0)} + H_f \qquad (22)$$

where subscript i stands for cl or q corresponding to the classical or quantum version of the description of the occupied microwave mode, respectively. $\mathbf{r}$ is the usual dipole operator, $\mathbf{E(r)}$ denotes the electric field operator at position $\mathbf{r}$ and H_f is the free field Hamiltonian. Both H_f and $\mathbf{E}$ can be expanded over modes of the field. While in [7], the expansion in sperical waves is used (which facilitates explicit transformation to the rotating frame), we expand the field into standard plane waves [3]. We normalize the modes per unit in $\mathbf{k}$ space (related to the frequency in atomic units by $\omega = \alpha k$ where $\alpha = e^2/\hbar c \simeq 1/137.036$ is the fine structure constant), then

$$\mathbf{E}(\mathbf{r}) = \sum_j \int \frac{\sqrt{\omega}}{2\pi} \left(a_j(\mathbf{k})\mathbf{e}_j \exp(i\mathbf{k} \cdot \mathbf{r}) + \text{h.c.} \right) \, d^3\mathbf{k}, \quad (23)$$

where h.c. stands for hermitian conjugation, $a_j(\mathbf{k})$ is the annihilation operator in the mode characterized by wavevector $\mathbf{k}$ and polarization $\mathbf{e}_j$ (the sum over j is over the two possible orthogonal polarizations). Note that in (22) the electric field operator is evaluated at origin, consistently with the dipole approximation assumed in this paper. Similarly the free field hamiltonian is

$$H_f = \sum_j \int \omega a_j(\mathbf{k})^\dagger a_j(\mathbf{k}) \, d^3\mathbf{k}. \qquad (24)$$

Now we can use the Fermi golden rule to calculate the decay rate due to spontaneous emission from an arbitrary initial state $|i\rangle$ of the atom dressed by the microwave field to any final state $|f\rangle$, taking into account the density of modes for the photons [31]. The result is:

$$\Gamma = \frac{4\alpha^3 \omega^3}{3} |(f|\mathbf{r}|i)|^2 \qquad (25)$$

where ω is the energy difference between the initial and final dressed states.

Spontaneous emission will take place between the various states belonging to ladders depicted in Fig. 1. In the process, the projection of the full angular momentum on the Oz axis is conserved. Here, this total angular momentum has three contributions: the atomic angular momentum m, the angular momentum N of the driving microwave photons (all in the same mode) and the angular momentum M in the other modes of the electromagnetic field. Hence, we have $\mathcal{M} = \mathcal{N} + M = N + m + M =$ constant. As spontaneous emission is here considered as a weak perturbation, it will simply induce transitions between eigenstates of the dressed atom, see Eq. (6), for which $\mathcal{N}$ is a good quantum number (see preceding section). In the dipole approximation, for a one-photon process, the atomic angular momentum m can change by at most one. When the spontaneous photon is emitted in an initially empty mode (which is the process we are interested in), the number of photons in the driving microwave mode N does not change. Hence, $\mathcal{N}$ changes by at most one, and the same is true (with opposite sign) for M.

Thus, in the dipole approximation, there are only three possible processes. If the spontaneous photon is emitted with σ^+ polarization, i.e. angular momentum $M = 1$, it will induce a transition from a state in a given $\mathcal{N}$ ladder to a state in the $\mathcal{N}' = \mathcal{N} - 1$ ladder. Similarly, for σ^- polarization, i.e. angular momentum $M = -1$, it will induce a transition from $\mathcal{N}$ to $\mathcal{N}' = \mathcal{N} + 1$ and for π polarization, $M = 0$ and $\mathcal{N}' = \mathcal{N}$. As all but one photon modes are initially empty, by spontaneous emission, the energy $\hbar\omega$ is gained in one mode and thus has to be lost by the dressed atom. Note that here and in the rest of this paper, we discuss the polarization of the various transitions using a quantization axis along the z axis, the direction of the propagation of the driving field. As explained below the spontaneous photons are mainly σ^+ polarized with respect to the z axis. Hence, the helicity of the photons (with respect to their direction of emission, i.e. the quantity measured in an experiment) is in general elliptical, depending on the direction of emission.

In summary, in the dressed atom picture as shown in Fig. 1, the only possibilities for emitting a spontaneous photon is to decay to a state with lower energy in the same ladder or in one of the two neighboring ladders. All other processes are forbidden. Note however that – in the absence of a further approximation – plenty of processes with all 3 possible polarizations should exist. However, as can be seen from Fig. 1, the σ^+ emission is clearly favored, since the $\mathcal{N} - 1$ ladder is shifted downwards with respect to the initial state by Ω. π is less favored and σ^- even less because of the upwards energy shift.

$\mathcal{N}+1$ $\mathcal{N}$ $\mathcal{N}-1$

σ^- polarization

$\Omega-\omega_+$

Ω

π polarization

$\Omega+\omega_-$

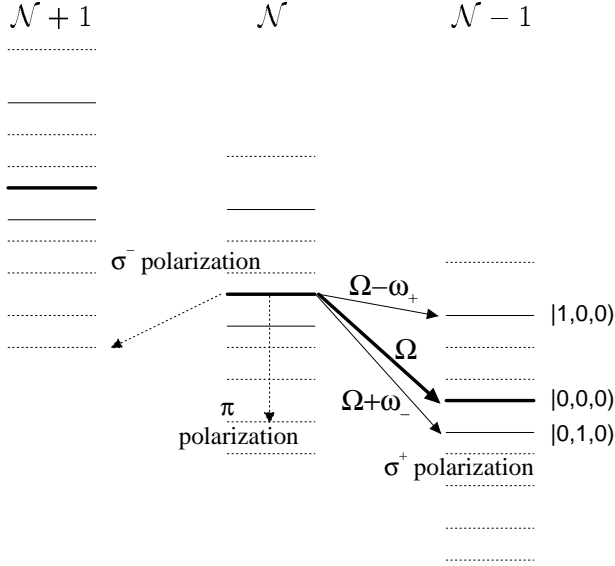σ^+ polarization

$|1,0,0)$

$|0,0,0)$

$|0,1,0)$

FIG. 1. Schematic energy levels of an hydrogen atom dressed by a microwave field of circular polarization, with Hamiltonian given by Eq. (6). The total angular momentum $\mathcal{N}$ (atom+field) is a constant of motion, so that the spectrum can be split in separate ladders. The consecutive ladders have the same structure, simply separated (in energy) by the microwave frequency Ω. We have here represented three consecutive ladders. If all ladders are taken into account, the spectrum is Ω-periodic and coincides with the Floquet spectrum of the classical Hamiltonian, Eq. (2). Alternatively, the spectrum in each ladder can be thought as the energy spectrum of the Hamiltonian in rotating frame, Eq.(3). Spontaneous emission of photons (represented by arrows in the figure) from a state in ladder $\mathcal{N}$ is only possible to another state lying at a *lower energy* in either ladder $\mathcal{N}-1$ (with σ^+ polarization), ladder $\mathcal{N}$ (with π polarization) or ladder $\mathcal{N}+1$ (with σ_- polarization). Hence, σ^+ transitions are favored. In the specific case of the ground non-dispersive wave packet $|0,0,0)$ in the harmonic approximation, only three transitions, represented by the solid arrows, are allowed at frequencies $\Omega-\omega_+$, Ω and $\Omega+\omega_-$, all of them being σ^+ polarized.

B. Spontaneous emission from a non-dispersive wave packet in the harmonic approximation

The general properties discussed in the preceding section are valid whatever initial state is chosen. When the initial state is a non-dispersive wave packet and when the scaled microwave field F_0 is sufficiently large, we can use the harmonic approximation described above. We will make this approximation throughout the rest of this section. The atomic dipole components $x \mp iy, z$ corresponding respectively to the photon polarizations $\sigma_\pm, \pi$ can be

expressed as functions of creation/annihilation operators in the normal modes, Eqs. (17)-(18): this is straightforward as all these operators are linear combinations of positions and momenta. The result is:

$$
\begin{aligned}
x \pm iy = \Omega^{-2/3} q^{-1/3} \\
+ \frac{1}{4\sqrt{\Omega}Q} \left(\frac{-3q + Q \pm (q-Q)\frac{\omega_+}{\Omega(1-q)}}{N_+} b_+^\dagger \right. \\
+ \frac{-3q + Q \mp (q-Q)\frac{\omega_+}{\Omega(1-q)}}{N_+} b_+ \\
+ \frac{3q + Q \pm (q+Q)\frac{\omega_-}{\Omega(1-q)}}{N_-} b_-^\dagger \\
\left. + \frac{3q + Q \mp (q+Q)\frac{\omega_-}{\Omega(1-q)}}{N_-} b_- \right)
\end{aligned} \tag{26}
$$

$$
z = \frac{q^{-1/4}}{\sqrt{2\Omega}}(b_z^\dagger + b_z) \tag{27}
$$

Hence, in the harmonic approximation, there are only very few transitions allowed. The selection rules are:

$$
\begin{array}{lll}
\Delta n_+ = 0 & \Delta n_- = 0, \pm 1 & \Delta n_z = 0 \text{ or} \\
\Delta n_+ = 0, \pm 1 & \Delta n_- = 0 & \Delta n_z = 0
\end{array} \tag{28}
$$

in σ^+ and σ_- polarizations and:

$$
\Delta n_+ = 0 \quad \Delta n_- = 0 \quad \Delta n_z = \pm 1 \tag{29}
$$

in π polarization.

This means that a given $|n_+, n_-, n_z; \mathcal{N})$ dressed eigenstate is connected to at most 2 states in the same $\mathcal{N}$ ladder and 5 states in each of the $\mathcal{N} \pm 1$ neighboring ladders. Among these 12 possible transitions, only those ending in a final state with lower energy are allowed in a spontaneous emission process. Using Eq. (21), we see that the energy loss is $\pm\omega_z$ for the 2 π-polarized transitions, $\Omega, \Omega \pm \omega_+, \Omega \pm \omega_-$ for the 5 σ_+ transitions and $-\Omega, -\Omega \pm \omega_+, -\Omega \pm \omega_-$ for the 5 σ_- transitions. From Eq. (15), we know that ω_+ and ω_- are both smaller than Ω. Hence, all 5 transitions in σ_- polarization are forbidden (by energy conservation) for spontaneous emission. On the other hand, all 5 transitions in σ_+ polarization are allowed. In π polarization, only the transition $\Delta n_z = -1$ is allowed by energy conservation. To summarize, in the harmonic approximation, a given $|n_+, n_-, n_z)$ atomic eigenstate can only emit spontaneous photons at the six frequencies $\omega_z, \Omega, \Omega \pm \omega_+, \Omega \pm \omega_-$ the first transition being π polarized, the 5 other ones having σ_+ polarization.

In the specific case where the initial state in the ground state wave packet $|0,0,0)$ with minimum values of the n_+, n_-, n_z quantum numbers and maximum localization around the equilibrium point, all transitions *decreasing* any of the n_+, n_-, n_z are obviously non-existent. Hence, we are left with only 3 possible transitions, all of them being σ_+ polarized:

- Transition from the ground state wave packet $|0,0,0;\mathcal{N})$ to its image $|0,0,0;\mathcal{N}-1)$ at microwave frequency Ω. While the atomic state is not changed, a photon of the driving microwave is elastically scattered into another mode at the same frequency, but with a different direction. In the language of quantum optics, this process can be viewed as the elastic component in the resonance fluorescence of the driven atom.

- Transition from $|0,0,0;\mathcal{N})$ to a $n_+ = 1$ dressed state $|1,0,0;\mathcal{N}-1)$ occurring at frequency $\Omega - \omega_+$. This is an inelastic component.

- Transition from $|0,0,0;\mathcal{N})$ to a $n_- = 1$ dressed state $|0,1,0;\mathcal{N}-1)$ occurring at frequency $\Omega + \omega_-$. This is also an inelastic component. Note that the difference on signs in the frequencies of the two inelastic components, $\Omega - \omega_+$ and $\Omega + \omega_-$ is directly related to the unusual - sign in the Hamiltonian, Eq. (20).

In the harmonic approximation, we can go beyond the above qualitative predictions and calculate the partial decay rates to the different final states analytically. To this end, we need the matrix elements of the dipole operators between the initial and final states. This is easily obtained using the expansion of the dipole operator on the creation/annihilation operators in the normal modes, Eq. (27), as the matrix elements of these operators between eigenstates are well known. We obtain:

$$(0,0,0|x-iy|0,0,0) = \Omega^{-2/3}q^{-1/3} \tag{30}$$

$$(1,0,0|x-iy|0,0,0) = \frac{-3q + Q - (q-Q)\frac{\omega_+}{\Omega(1-q)}}{4\sqrt{\Omega}QN_+} \tag{31}$$

$$(0,1,0|x-iy|0,0,0) = \frac{3q + Q - (q+Q)\frac{\omega_-}{\Omega(1-q)}}{4\sqrt{\Omega}QN_-}. \tag{32}$$

This finally makes it possible to calculate the decay rates of the 3 spontaneous emission processes, using Eq. (25):

$$\Gamma(\Omega) = \frac{2\alpha^3\Omega^{5/3}q^{-2/3}}{3} \tag{33}$$

for the elastic component,

$$\Gamma(\Omega - \omega_+) = \frac{2\alpha^3(\Omega - \omega_+)^3}{3}|(1,0,0|x-iy|0,0,0)|^2 \tag{34}$$

and

$$\Gamma(\Omega + \omega_-) = \frac{2\alpha^3(\Omega + \omega_-)^3}{3}|(0,1,0|x-iy|0,0,0)|^2 \tag{35}$$

for the two inelastic components. The various quantities in these formula are related to the microwave frequency Ω and to the scaled parameter q (itself related to the microwave amplitude by Eqs. (10) and (11)) by Eqs. (15)

and (32). For a fixed classical dynamics (fixed q i.e. fixed scaled microwave amplitude F_0), the elastic decay rate scales as $\Omega^{5/3} = n_0^{-5}$ as does the decay rate of a circular state while the inelastic decay rates scale differently as $\Omega^2 = n_0^{-6}$. The results can be converted to usual units by multiplying them by $me^4/\hbar^3 \simeq 4.13 \times 10^{16} \text{ s}^{-1}$.

In Fig. 2, we show the frequencies, squared dipole matrix elements and decay rates (in linear and logarithmic scales) of the three components as a function of the scaled microwave field for principal quantum number $n_0 = 60$, the corresponding microwave frequency being $\Omega/2\pi = 30.46$ GHz. The first thing to note is obviously the dominance of the elastic component at the microwave frequency. About 80% of the spontaneous photons emitted by the system are at the microwave frequency. Elastic scattering of the microwave photons to other modes (different direction) is by far the dominant process. The radiation diagram of these photons has the standard dipolar shape (this is identical for the three components as they are all due to the $x - iy$ dipole operator). This dominance is easily understood from the different scalings of the three decay rates, Eqs. (33)-(35) discussed above. The dipole matrix element $(0,0,0|x-iy|0,0,0)$ measures the static dipole, i.e. the position of the center of the non-dispersive wave packet. It is thus – in the harmonic approximation - equal to x_e, the coordinate of the classical fixed point, of the order of n_0^2. In contrast, the non diagonal matrix elements $(0,1,0|x-iy|0,0,0)$ or $(1,0,0|x-iy|0,0,0)$ measure how the excited states explore the vicinity of the fixed point. They are of the order of $n_0^{3/2}$, i.e. about $\sqrt{n_0}$ times smaller. This is why the corresponding decay rates are much smaller. Because of the different scaling properties of the decay rates, it is easy to check that a change in the value of n_0 gives similar plots for the decay rates of the inelastic components as a function of the scaled microwave field (they are just multiplied by a uniform numerical factor) while the elastic component is relatively increased in proportion with n_0. In other words, in the semiclassical limit $n_0 \to \infty$, the inelastic components asymptotically vanish.

Another important observation is that, although the two inelastic components have dipole matrix elements of comparable magnitude (see Fig. 2b), the decay rate of the $\Omega - \omega_+$ component is several orders of magnitude smaller than the decay rate of the $\Omega + \omega_-$ component, see Fig. 2c. This is entirely due to the ω^3 factor representing the density of electromagnetic mode. In practice, the $\Omega - \omega_+$ component will be very difficult to detect.

Finally, when the microwave field is sufficiently small, the various quantities can be expanded in powers of the scaled field. In this limit (see also Fig. 2d), the $\Omega - \omega_+$ decay rate goes to zero as F_0^3 whereas the $\Omega + \omega_-$ decay rate increases like $1/\sqrt{F_0}$ and the elastic component Ω keeps a constant decay rate. Of course, such a divergence in clearly unphysical and simply indicates the breakdown of the harmonic approximation. At lower field, the pendulum approximation of the Hamiltonian [21] has to be

used, restoring finite decay rates, roughly equally shared by the $\Omega + \omega_-$ and Ω components.
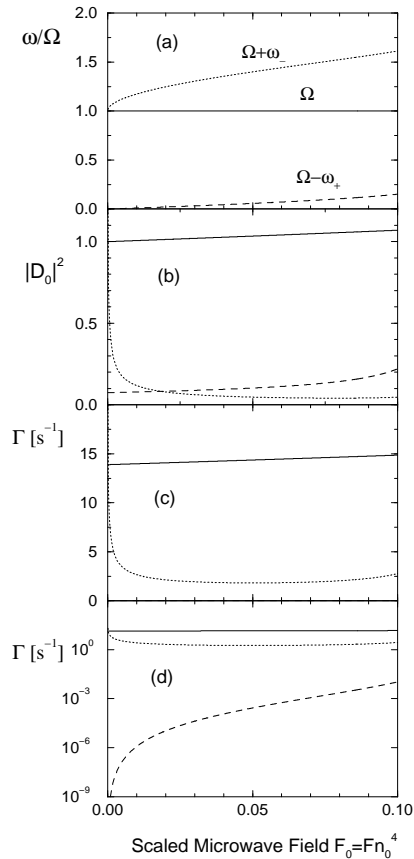

FIG. 2. Analytic predictions for the spectrum of spontaneous photons emitted by a non-dispersive wave packet $|0,0,0\rangle$ of the hydrogen atom in a cicularly polarized microwave of frequency $\Omega/2\pi = 30.46$ GHz corresponding to a principal quantum number $n_0 = 60$, as a function of the scaled microwave field amplitude. Solid line corresponds to elastic scattering of the microwave photons, the dashed and dotted lines correspond to inelastic processes. (a): Frequencies of the emitted photons. (b): Squares of the dipole matrix elements involved in the 3 transitions. The elastic component is largely dominant. (c): Decay rates along the 3 transitions. Because of the rapid increase in the density of electromagnetic modes with frequency, the $\Omega - \omega_+$ component is almost invisible. (d): Same as (c) on a logarithmic scale. The decay rates of the $\Omega - \omega_+$, Ω and $\Omega + \omega_-$ components roughly scale as $F_0^3 n_0^{-6}$, n_0^{-5} and $F_0^{-1/2} n_0^{-6}$, respectively.

IV. COMPARISON WITH EARLIER RESULTS

The former approach to spontaneous emission of the non-dispersive wave packets [7] has been carried out in the rotating frame using the Hamiltonian Eq.(3), or rather its generalization which includes also other non

occupied electromagnetic modes. Using the notion of rotational frequency shift found previously by the same authors [13], the frequency of the emission in the laboratory frame has been found. A single frequency, since the authors considered the emission at $\Omega + \omega_-$ only assuming, incorrectly, that spontaneous emission of a photon in the rotating frame must lead to a decrease of the energy (as it happens in the laboratory frame).

To see the fallacy of this argument, consider the standard spontaneous decay of the circular $|n+1, n, n\rangle$ hydrogenic state in the absence of any driving electromagnetic field. In the dipole approximation, it can decay spontaneously only to the $|n, n-1, n-1\rangle$ circular state. Consider the same situation in the frame rotating at frequency Ω. The Hamiltonian in the rotating frame is obtained from Eq. (3) with $F = 0$. The energy of the initial state in the rotating frame is then $E_i = -1/2(n+1)^2 - \Omega n$ while the energy of the final state $E_f = -1/2n^2 - \Omega(n-1)$ is greater than E_i provided $\Omega > (n + 1/2)/(n(n + 1))^2 \approx 1/n^3$. Hence, emission of the spontaneous σ^+ polarized photon *increases* the energy in the rotating frame.

The same situation occurs for the standard toy model of quantum optics: the two-level atom interacting with a quasi-resonant monochromatic field, treated in the rotating wave approximation. In the "rotating frame" [23], spontaneous emission does not occur only from the upper to the lower state of the (Autler-Townes) doublet. This particular line (upper to lower state in the doublet) occurs at the frequency of the driving field plus the Rabi frequency. But the emission may also take place from the lower to the upper state of the doublet (at driving frequency minus the Rabi frequency) as well as from lower (upper) state to "itself". In effect the resonance fluorescence spectrum has the famous Mollow triplet structure [32,3] with the dominant elastic component in the middle.

This discussion shows that the rotating frame approach leads to quite counter intuitive picture. By comparison, the dressed state picture presented in the previous section seems to be quite intuitive. One may easily verify that extending the calculation presented in [7] to include the lines emitted at the microwave frequency Ω (which corresponds in the rotating frame to the emission from the wave packet state to itself) as well as at $\Omega - \omega_+$ leads to the same results as those obtained in the dressed atom picture above [33]. This shows that both approaches lead, as it should be, to the same spontaneous emission rates.

From a practical point of view, as the decay at $\Omega - \omega_+$ is much slower than the decay at $\Omega + \omega_-$ (see discussion above), the numbers given in [7] can be considered correct [34] if elastic scattering does not have to be taken into account. A non-trivial question is whether elastic scattering of photons has to considered as a decay process or not. In our opinion, this is simply a question not properly formulated. Thinking resonance fluorescence in terms of decay is probably inadequate as the system is continuously fed with external photons. If one measures the

population in the ground state wave packet $|0,0,0\rangle$, this quantity will certainly decay because of inelastic components only. If one measures the spectrum radiated by the atom, the answer is completely different as the elastic component dominates. Also, if one considers the widths of the various components, the elastic process will come into the game as it destroys the phase coherence of the initial wave packet and consequently broadens the line. A general discussion of such effects and more generally of resonance fluorescence in the framework of quantum optics can be found in [32].

Let us also note that the "classical" emission rate of an electron moving on a circular orbit presented in [7] is incorrect. A classical charge moving on a circular orbit with frequency Ω must emit photons at the same frequency. In our opinion, the only meaningful quantity is the decay rate of the *energy* in the atomic system. Classically, it is proportional to the square of the acceleration and, for the classical periodic orbit at the microwave frequency, it is in atomic units [35]:

$$\frac{dE}{dt} = \frac{2\alpha^3 \Omega^{8/3} q^{-2/3}}{3} \qquad (36)$$

Considering that, in the semiclassical limit $n_0 \to \infty$, the system radiates essentially photons as frequency Ω, we immediately see that this classical rate *exactly* (i.e. without additional numerical factor of the order of one) coincides with the quantum decay rate of energy deduced from Eq. (33). It also exactly corresponds to the decay rate of atomic circular states, as given in e.g. Ref. [36], in the limit of vanishing microwave field $q \to 1$. In [7], the decay rate in the $\Omega + \omega_-$ component is incorrectly compared to the relative decrease of the classical energy in time. The first quantity – as discussed above – is roughly n_0 times smaller than the elastic decay rate while the relative decrease of classical energy in time is also about n_0 times smaller than the rate of emission of photons because the energy of a radiated photon is about n_0 times smaller than the total energy. The two n_0 dependences cancel out, but not the numerical factors in front. This explains why a false classical-quantum correspondence (within constant numerical factors) is observed in [7]. Both classical and quantum decay rates are – in our opinion – underestimated by a $\simeq n_0$ factor.

V. HOW GOOD IS THE HARMONIC APPROXIMATION ?

From the beginning of investigations on non-dispersive wave packets in the hydrogen atom driven by a circularly polarized microwave, there has been a controversy regarding the accuracy of the harmonic approximation used when introducing the wavepackets as Gaussians centered around the equilibrium point [1], see Section II. Firstly in a Comment [15] and later in a number of papers [16–20] Farrelly and coworkers claimed that "highly

anharmonic nature of the effective potential in the vicinity of the equilibrium point will cause any such putative coherent state to disperse .." unless the wave packet is built from states of unrealistically high quantum number ($n_0 > 200$). They have shown that an additional static magnetic field applied in the direction of microwave propagation (Oz axis) allows to relatively decrease the importance of the anharmonic terms in the expansion around the equilibrium point. Further they claim that "because these extrema are locally harmonic their vacuum states are truly coherent states in the original sense of Schrödinger" [20] (by extrema one should understand global equilibrium points).

Our understanding of the phenomenon is slightly different. While Farrelly and coworkers stress that the Gaussian shape of the wave packet is essential for non spreading time evolution in the locally harmonic potential, we adopt a bit less narrow-minded point of view. We have shown [4] that, even if the harmonic approximation is only partially valid, there will be a one-to-one correspondence between the quasienergy states of the approximate harmonic Hamiltonian, Eq. (20), and the full Hamiltonian in the rotating frame, Eq. (3). In particular, the ground state of the local harmonic approximation (referred to above as ground state wave packet) is a good approximation to an *exact* eigenstate of the full problem (or via the unitary transformation to the corresponding Floquet or dressed state). We refer to this exact eigenstate of the full atom+microwave field problem as the non-dispersive wave packet since:

- it is a linear combination of atomic states;

- it is centered at the stable equilibrium point in the rotating frame, thus it follows the classical circular periodic orbit of the problem in the laboratory frame;

thus fulfilling standard textbook criteria for "a wave packet". Being an eigenstate of $H_{\rm rot}$, Eq. (3), the state is rigorously non spreading provided we neglect the coupling to the continuum – i.e. the possibility of ionization. We have shown [4–6] that typical lifetimes for the ionization exceed many thousands of Kepler periods (reaching up to 10^6 Kepler periods for $n_0 \approx 60$ or more).

In our understanding of the phenomenon, the validity of the harmonic approximation is not crucial for the very existence of non-dispersive wave packets in the Hydrogen atom driven by a circularly polarized microwave. Moreover, the phenomenon is more general and, as mentioned in the introduction, may be found in other systems such as atoms driven by a linearly polarized microwave [2] or one-dimensional models [25] supporting stable classical 1:1 resonance islands. In the general case, the wave packet preserves its shape at intervals equal to the period of the external driving, only in the circular polarization problem (due to the additional symmetry which allows for the complete removal of the time-dependence) the wave packet shape is the same at all times. There is no

reason to restrict the notion of non-dispersive (or non spreading) wave packet to a single particular situation which can be made almost fully harmonic. One should also remember that even in the presence of a magnetic field, a case favored by Farrelly and coworkers, the potential is also only approximately harmonic, and Gaussian wavepackets will eventually disperse. While wave packets in our sense, being single dressed states, will not. In any case, the addition of a magnetic field does not change the nature of the problem, it may just keep some anharmonic terms smaller.

While not necessary for the very existence of non-dispersive wave packets, the harmonic approximation is useful for analytic evaluation of different properties, such as spontaneous emission discussed above. In view of Farrelly and coworkers remarks, it would seem that the results obtained in the previous section are incorrect. The simplest way to check this statement is to evaluate the "exact" spontaneous emission spectrum from the ground quasienergy eigenstate of the full Hamiltonian.

The presence of the anharmonic terms in the full Hamiltonian has to manifest itself in the break-up of the selection rules discussed in Section II. Instead of three possible lines in the σ^+ emission, one should expect a multitude of lines. Using the harmonic level assignment, the $|0, 0, 0; \mathcal{N}\rangle$ wave packet may then decay to e.g. states approximated by $|i, j, k; \mathcal{N} - 1\rangle$ with i, j, k being larger than unity (with k even since σ^+ dipole element may link states of same z-parity only). Also, due to couplings introduced by nonlinear terms, spontaneous emission with other (i.e. σ_- or π) polarizations becomes possible.

Thus, the spontaneous emission spectrum provides a useful test of the validity of the harmonic approximation. Having at our disposal the computer code which allows reliably to find the exact quasienergy states of the problem [4–6], it has been sufficient to evaluate dipole matrix elements between the exact quasienergy states. The resulting squared dipole moments are shown in Fig. 3 together with the corresponding squared dipoles as given by the harmonic approximation, Eq. (32), presented as three crosses. Observe that, the harmonic approximation works quite well. The selection rules are well preserved, the frequencies of the emission (line positions) agree very well, the dipole values (heights of the lines) agree within 10% for all three lines. This shows that the harmonic approximation is quite good, *in the absence of any magnetic field,* not only for the ground state wave packet, $|0, 0, 0\rangle$ but also for primary ω_+ [state $|1, 0, 0\rangle$] and ω_- [state $|0, 1, 0\rangle$] excitations. Observe that the height of the peak at Ω is proportional to $|\langle 0, 0, 0|x - iy|0, 0, 0\rangle|^2$ involving the wavefunction of the ground state wave packet only. The sidebands, corresponding to transitions to excited wave packets provide independent tests of the corresponding wavefunctions.

Fig. 4 shows the corresponding spontaneous emission rates. The component at $\Omega - \omega_+$ is not visible due to the low frequency of the emission. On the other hand, the spectrum at high frequencies is enhanced by cubic power

of the frequency factor coming from the photonic density of states. Here, careful inspection of the spectrum corresponding to exact eigenstates reveals a tiny peak centered at $\Omega + 2\omega_-$ (indicated by an arrow in the figure). Such a transition is forbidden in the harmonic approximation, its presence indicates that the harmonic approximation is not exact. Still, the smallness of anharmonic terms strongly indicates that the harmonic approximation works well, at least for the typical value $n_0 = 60$.

If one considers lower n_0 (say around $n_0 = 30$) when the effective $\hbar$ is bigger, the anharmonic terms in the Hamiltonian become more important and the spontaneous emission spectrum contains other lines, at $\Omega + k\omega_-$ with $k \geq 2$ or combinations, e.g. at $\Omega + 2\omega_- - \omega_+$. This nicely correlates with our study of ionization mechanism [6]. Starting around $n_0 = 40$, ionization clearly occurs via a chaos assisted tunneling mechanism (obeying exponential behavior with $\hbar$ change) with clear deviations from such a behavior for lower n_0.

Similarly, much weaker microwave amplitudes (say $F_0 = 0.005$) when the resonance island is so small that the quantum eigenstate does not fit into it, lead to a similar behavior of the spontaneous emission spectrum - the presence of overtones. Still we have checked that in quite a broad range of $0.01 < F_0 < 0.06$, the harmonic approximation works similarly to the case shown in Fig. 3 and Fig. 4, provided no accidental avoided crossing with other Floquet states occurs [37].

While the calculations presented in this paper are consistently done at the lowest order of the harmonic approximation using Cartesian coordinates, similar calculations can be done using, for example, the spherical coordinates [11,12]. Consistently, the results at lowest order are of course identical. However, it is known that – within the harmonic approximation – the wavefunction in spherical coordinates is a better approximation to the "exact" wavefunction than the corresponding one in Cartesian coordinates, as it incorporates some of the anharmonic terms; the price to pay is the breaking of the selection rules, Eqs. (28)-(29). Hence, the transition rates calculated using harmonic wavefunctions in spherical coordinates incorporate already some of the higher order terms and might be closer to the "exact" results. Note, however that if the calculation is performed consistently at any order in perturbation theory (the anharmonic terms being the perturbation), the results should of course be independent of the choice of coordinates.
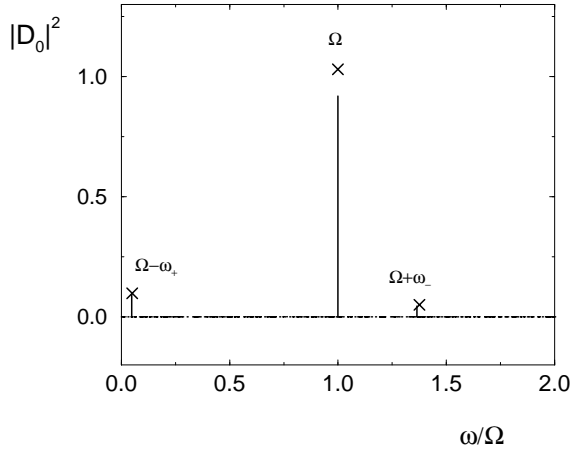
FIG. 3. Single photon spontaneous emission from the non-dispersive wave packet of the hydrogen atom in a cicularly polarized microwave of frequency $\Omega = 1/60^3$ (in atomic units) corresponding to a principal quantum number $n_0 = 60$, and scaled amplitude $F_0 = 0.04445881$ (corresponding to $q = 0.9562$). We have here represented the square of the scaled dipole matrix element $D_0 = D/n_0^2$ connecting the initial $|0,0,0\rangle$ wave packet to the final state as a function of the scaled energy difference (hence, ratio of the frequency of the emitted photon to the microwave frequency Ω) between the two states. The stick spectrum is the "exact" result as obtained from a numerical diagonalization and the crosses represent the analytic result obtained using the harmonic approximation, Eq. (32). There are three dominant lines at frequencies $\Omega - \omega_+$, Ω and $\Omega + \omega_-$, all of them σ^+ polarized. The strongest line (by a factor proportional to n_0) is the elastic scattering of the microwave photons by the dressed atom. The other transitions (as well as transitions with σ_- or π polarizations) are negligible, the corresponding sticks have practically zero heights (yielding a broken line build of individual sticks each representing one of the over 400 Floquet final states), which proves the validity of the harmonic approximation.
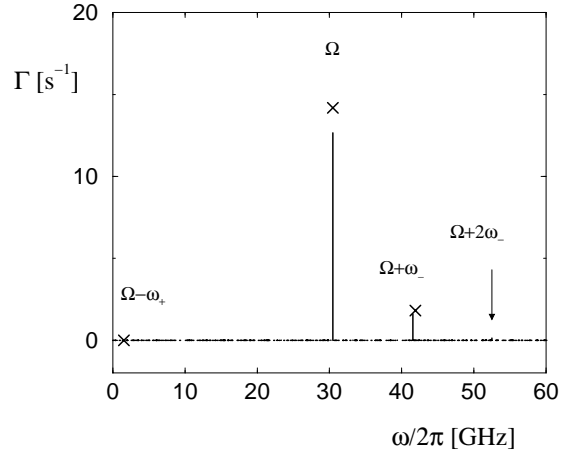

FIG. 4. Same as figure 3, but for the decay rates along the different transitions and in S.I. units. The density of modes of the electromagnetic wave introduces an additional ω^3 factor which completely kills the transition at frequency $\Omega - \omega_+$, invisible in the figure. One can see a small line at frequency approximately $\Omega + 2\omega_-$ (see arrow), an indication of a weak breakdown of the harmonic approximation.

VI. CONCLUSIONS

It is worth stressing again that, in the semiclassical limit (small Ω or large n_0), the dominant contribution to the spontaneous emission comes from the elastic component at frequency Ω which *does not* destroy the wave packet but rather leads to the decoherence in its time-evolution. The wave packet converts the energy of the driving photon into a spontaneously emitted photon, moving at the same time on the circular orbit.

From a practical point of view, the spontaneous emission rates are several orders of magnitude smaller than the ionization rate in a typical situation $n_0 = 60$ and $F_0 = 0.05$ [6], and thus unlikely to be observable. However, this ratio can be made smaller by increasing n_0. Indeed, as shown in [6], the typical ionization rate decreases *exponentially* with n_0 (because it is due a chaos assisted tunneling process) while the spontaneous decay rates decrease *algebraically* with n_0. For very large n_0, spontaneous emission will eventually become the dominant process. For $F_0 = 0.05$, the cross-over should take place around $n_0 = 200$ which makes it very difficult to observe experimentally. Another interesting possibility is to decrease the scaled field F_0 so that the ionization rate drops considerably. According to numbers shown in [6], the cross-over for $n_0 = 60$ should take place around $F_0 = 0.03$ in a regime where the harmonic approximation is still valid (the results are not shown since they are very similar to those depicted in Fig. 4). In any case,

the total decay rate is of the order of few tens of photons per second, making the experimental observation rather difficult.

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[23] In the context of quantum optics of a two-level system, the rotating wave approximation makes it possible to define a time-dependent unitary transformation producing a static Hamiltonian. As there is a formal analogy, this is known as passing to the "rotating frame" although this is in general not a purely geometrical transformation. Only in the case of a circularly polarized field do the two acceptions of "rotating frame" coincide.

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[30] We have chosen the length gauge for the description of the empty modes of the electromagnetic field. Of course, one could use a different gauge, such as the velocity gauge. The matrix elements coming into play are different, but we checked that the decay rates of the various spontaneous transitions are – as they should – gauge independent. Similarly, we could use another gauge to describe the driving microwave field. This produces a different Floquet Hamiltonian and a different static Hamiltonian in the rotating frame, hence different creation/annihilation operators for the normal modes. However, the energy levels of the dressed atom – and consequently the frequencies of the normal modes – do not depend on the choice of gauge. The same is true for the decay rates. The "exact" computations shown in this paper have been performed in velocity gauge.

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magnetic modes in free space. The decay rates can be substantially modified in the presence of a cavity, as could be used in a real experiment [8].

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[34] Note however that the spontaneous decay rate per Kepler period as given in [7] at the end of page 3630 has a 2π factor wrongly placed. It is thus "only" 4 orders of magnitude smaller than the ionization rate.

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